

Finite-Temperature Charge-Ordering Transition and Fluctuation Effects in Quasi-One-Dimensional Electron Systems at Quarter Filling

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Finite-temperature charge-ordering phase transition in quasi one-dimensional (1D) molecular conductors is investigated theoretically, based on a quasi 1D extended Hubbard model at quarter filling with interchain Coulomb repulsion V_{\perp} . The interchain term is treated within mean-field approximation whereas the 1D fluctuations in the chains are fully taken into account by the bosonization theory. Three regions are found depending on how the charge ordered state appears at finite temperature when V_{\perp} is introduced: (i) weak-coupling region where the system transforms from a metal to a charge ordered insulator with finite transition temperature at a finite critical value of V_{\perp} , (ii) an intermediate region where this transition occurs by infinitesimal V_{\perp} due to the stability of inherent 1D fluctuation, and (iii) strong-coupling region where the charge ordered state is realized already in the purely 1D case, of which the transition temperature becomes finite with infinitesimal V_{\perp} . Analytical formula for the V_{\perp} dependence of the transition temperature is derived for each region.

KEYWORDS: charge-ordering transition, quarter filling, quasi one-dimension, extended Hubbard model

Charge-ordering (CO) phenomenon has been one of the central research subjects in quasi one-dimensional (1D) organic 2:1 salts such as $(\text{DCNQI})_2\text{X}^{1-3}$ and $(\text{TMTTF})_2\text{X}^{4-6}$ (X : monovalent counter ion). Some members of them having a *quarter-filled* electron or hole band exhibit insulating behavior due to CO. For example, in $(\text{DI-DCNQI})_2\text{Ag}$, an NMR measurement clarified the CO phase transition at $T_{\text{CO}} = 220$ K,¹ above which non-metallic behavior is already seen even from room temperature interpreted as due to CO fluctuation.³ In $(\text{TMTTF})_2\text{X}$, CO is found by NMR as well^{4,6} and the dielectric constant shows a divergence toward T_{CO} , where $T_{\text{CO}} = 70 \sim 160$ K depending on X .⁵

Theoretical studies showed that a minimal model for describing such CO is the 1D quarter-filled extended Hubbard model (EHM) with not only the on-site Coulomb repulsion U but also the intersite repulsion V .⁷ This model has been intensively studied and the *ground-state* properties have been revealed. Using different numerical techniques, the $T = 0$ phase diagram on the (U, V) plane has been obtained with high accuracy.⁸ The CO insulator (COI) appears in the large (U, V) region. Analytical approaches based on the bosonization theory clarified the mechanism of the transition from a Tomonaga-Luttinger liquid (TLL) metallic phase to the COI.⁹⁻¹²

It is useful to consider the TLL parameter K_{ρ} depending on (U, V) in the metallic phase of this 1D EHM. It takes $K_{\rho} > 1/4$ and approaches to $K_{\rho} = 1/4$ at the boundary between the TLL and the COI. Based on the TLL theory, CO fluctuation is known to develop as $\chi_{\text{CO}}(T) \propto T^{4K_{\rho}-2}$, which implies that for $1/2 > K_{\rho} > 1/4$ it diverges at low temperatures. This divergence does not immediately point to the appearance of long range order of CO in the purely 1D systems. The COI is achieved for larger (U, V) , only at $T = 0$. These

features are schematically illustrated in Fig. 1 (a). When U is fixed at a large value (e.g., $U \gtrsim 4t$; see Fig. 2 later) and V is increased, K_{ρ} decreases, and the system transforms through three regions bounded by $V = V_f^{1\text{D}}$ and $V = V_c^{1\text{D}}$ defined by the values where $K_{\rho} = 1/2$ and $1/4$, respectively. The system is the TLL for (i) $V < V_f^{1\text{D}}$ and (ii) $V_f^{1\text{D}} < V < V_c^{1\text{D}}$ where in the latter CO fluctuation develops at low- T , and it is the COI for (iii) $V_c^{1\text{D}} < V$.

In order to examine the *finite-temperature* CO transition in the quasi 1D materials, in addition to such 1D fluctuation we need to take into account the dimensionality effect by the interchain coupling. In these systems two types of interchain coupling exist in the electronic sector; one is the single-particle interchain hopping and the other is the Coulomb interaction between electrons in the different chains. In this letter, we consider the latter and discuss the critical temperature of CO, T_{CO} , by treating the interchain interaction within the mean-field approximation, whereas taking full account of the 1D fluctuations in the chains by the bosonization theory. We will find distinct behavior for the three regions defined above : in region (i) the system transforms from TLL metal to

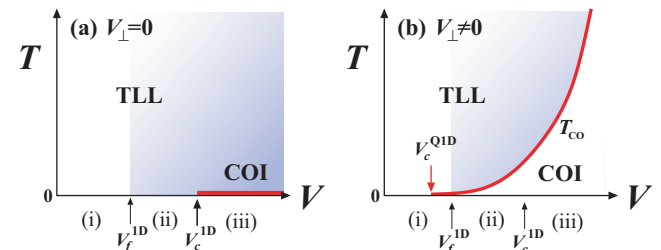


Fig. 1. Schematic illustration of the finite-temperature phase diagram for purely 1D case (a) and quasi-1D case with finite V_{\perp} (b), where the CO fluctuation develops in the shaded regions. Here V_c^{Q1D} express the critical value of V for the COI appearing in the quasi-1D case. The quantities $V_c^{1\text{D}}$ and $V_f^{1\text{D}}$, and the regions, (i), (ii) and (iii) are defined in the text.

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COI with finite T_{CO} at a finite critical value of V_{\perp} , in (ii) this transition occurs by infinitesimal V_{\perp} , and in (iii) COI in the purely 1D case always has finite T_{CO} when V_{\perp} is turned on. A schematic phase diagram in the presence of V_{\perp} is summarized in Fig. 1 (b).

Our Hamiltonian is given by $H_{\text{Q1D}} = \sum_j H_{\text{1D}}^j + H_{\perp}$. Here H_{1D}^j represents the j -th isolated extended Hubbard chain:

$$H_{\text{1D}}^j = -t \sum_{i,s} \left(c_{i,j,s}^{\dagger} c_{i+1,j,s} + \text{h.c.} \right) + U \sum_i n_{i,j,\uparrow} n_{i,j,\downarrow} + V \sum_i n_{i,j} n_{i+1,j}, \quad (1)$$

and H_{\perp} expresses the interchain coupling:

$$H_{\perp} = V_{\perp} \sum_{i,\langle j,j' \rangle} n_{i,j} n_{i,j'}, \quad (2)$$

where $c_{i,j,s}^{\dagger}$ is the creation operator of an electron with spin $s = \uparrow/\downarrow$ at the i -th site in the j -th chain, $n_{i,j,s} = c_{i,j,s}^{\dagger} c_{i,j,s} - 1/4$ and $n_{i,j} = n_{i,j,\uparrow} + n_{i,j,\downarrow}$. In eq.(2), $\langle j, j' \rangle$ denotes the pair of adjacent chains and the strength of interchain Coulomb repulsion is expressed by V_{\perp} . The number of adjacent chains, z , for (DCNQI) $_2X$ and (TMTTF) $_2X$ would be $z = 4$ and $z = 2$ judging from the crystal structures, although their interchain networks are rather complicated, as we will discuss later. We do not take into account the interchain hopping which should not yield qualitative changes to the present analysis at the temperature above the crossover energy scale due to the interchain hopping.¹³

We treat eq. (2) in the interchain mean-field approach,¹⁴ which is known to be effective in the weak V_{\perp} region, by considering the Wigner-crystal-type CO pattern stabilized in the 1D EHM. We assume the CO pattern to be anti-phase between adjacent chains, which is naturally expected to gain the V_{\perp} term. The resulting effective 1D Hamiltonian is written as

$$H = -t \sum_{i,s} \left(c_{i,s}^{\dagger} c_{i+1,s} + \text{h.c.} \right) + U \sum_i n_{i,\uparrow} n_{i,\downarrow} + V \sum_i n_i n_{i+1} + z V_{\perp} n \sum_i (-1)^i n_i + z N V_{\perp} n^2 / 2, \quad (3)$$

where the chain index j is suppressed and N is the total number of sites in a chain. The amplitude of CO is written as n .

To obtain a qualitative insight of this model we first discuss the $U \rightarrow \infty$ limit, where some exact results for the 1D model based on the Bethe ansatz can be used. In this case, we can neglect the spin degree of freedom which acts freely, and the charge degree of freedom is reduced to a half-filled spinless fermion model.¹⁵

$$H_{U \rightarrow \infty} = -t \sum_i (d_i^{\dagger} d_{i+1} + \text{h.c.}) + V \sum_i \tilde{n}_i \tilde{n}_{i+1} + z V_{\perp} n \sum_i (-1)^i \tilde{n}_i + z N V_{\perp} n^2 / 2, \quad (4)$$

where the creation operator of the spinless fermion is expressed by d_i^{\dagger} and $\tilde{n}_i \equiv d_i^{\dagger} d_i - 1/2$. This model is equivalent

to the $S = 1/2$ XXZ spin-chain coupled by the interchain exchange interaction $J_{\perp} (= V_{\perp})$ treated in the interchain mean-field approach.^{16,17} Several authors have performed such approach to the Heisenberg limit (i.e., $V = 2t$).¹⁸ Bhaseen and Tsvelik discussed a nontrivial spectrum of eq. (4) in a parameter region $V \geq 2t$,¹⁹ where the system is the COI and has a finite excitation gap even for $V_{\perp} = 0$.¹⁵ We restrict ourselves to a parameter region $0 < V \leq 2t$, where the system is known to be metallic if $V_{\perp} = 0$, and show that infinitesimally small V_{\perp} makes the system insulating and gives rise to finite T_{CO} .

We apply the bosonization method to eq. (4). The density operator is given by $\tilde{n}_i = a(\sqrt{2}\pi)^{-1} \partial_x \theta(x) - \pi^{-1}(-1)^i \sin \sqrt{2}\theta(x)$ with $x = ia$ and a being the lattice constant. This parameter region $0 < V \leq 2t$ corresponds to region (ii) with $1/2 > K_{\rho} \geq 1/4$ defined above, since it is known that in the 1D Hubbard model at $U \rightarrow \infty$ ($V = 0$), K_{ρ} takes a universal value $K_{\rho} \rightarrow 1/2$,²⁰ and at $V = 2t$, the CO transition indicates $K_{\rho} = 1/4$.

In the bosonized form of the Hamiltonian, there appears a nonlinear term, $V/(2\pi^2 a) \cos 2\sqrt{2}\theta$, which becomes irrelevant but has an effect to renormalize the velocity, the TLL parameter and the prefactor of the bosonized density operator. By taking into account these effects properly, the density operator is given by $\tilde{n}_i = a(\sqrt{2}\pi)^{-1} \partial_x \theta(x) - c\pi^{-1}(-1)^j \sin \sqrt{2}\theta(x)$, where c is a nonuniversal constant depending on V/t and its exact value has been suggested.²¹ We will use the renormalized velocity and the TLL parameter, and neglect the $\cos 2\sqrt{2}\theta$ term. After this procedure, we switch on the V_{\perp} term, and then the effective Hamiltonian density reads

$$\mathcal{H}_{U \rightarrow \infty}^{\text{eff}} = \frac{v}{4\pi} \left[\frac{1}{2K_{\rho}} (\partial_x \theta)^2 + 2K_{\rho} (\partial_x \phi)^2 \right] - \frac{g_{\perp}}{2\pi^2 a^2} \sin \sqrt{2}\theta + \frac{z}{2a} V_{\perp} n^2, \quad (5)$$

where $v = \pi t a \sqrt{1 - (V/2t)^2} / \arccos(V/2t)$, $K_{\rho} = [4 - (4/\pi) \cos^{-1}(V/2t)]^{-1}$, and $g_{\perp} = 2\pi c z V_{\perp} a n$. The TLL parameter K_{ρ} determines the nonuniversal constant $c = c(K_{\rho})$.

Since the g_{\perp} term in eq. (5) is a relevant perturbation, when $n \neq 0$ the system is not the TLL but has an excitation gap,²²

$$m(n) = \frac{v}{a} \left[\frac{g_{\perp}}{4\pi^2 v \kappa \left(\frac{K_{\rho}}{1-K_{\rho}} \right)} \right]^{1/(2-2K_{\rho})}, \quad (6)$$

where $\kappa(p) \equiv \pi^{-1}(\pi/4)^{1/(p+1)} \Gamma(p/(p+1)) \Gamma^{-1}(1/(p+1)) [\Gamma((p+1)/2) \Gamma^{-1}(p/2)]^{2/(p+1)}$. The ground-state energy per site is given by²²

$$E(n) = -\frac{a}{4v} m^2(n) \tan \left(\frac{\pi K_{\rho}}{2 - 2K_{\rho}} \right) + z V_{\perp} n^2 / 2. \quad (7)$$

The CO amplitude n is determined from the condition to minimize eq. (7). The optimized magnitude n is given by

$$n = \frac{1}{2} \left[\frac{\tan(\pi K_{\rho}/(2 - 2K_{\rho}))}{2 - 2K_{\rho}} \right]^{\frac{1-K_{\rho}}{1-2K_{\rho}}} \times \left[\frac{c(K_{\rho})}{2\pi \kappa \left(\frac{K_{\rho}}{1-K_{\rho}} \right)} \right]^{\frac{1}{1-2K_{\rho}}} \left(\frac{z V_{\perp} a}{2v} \right)^{K_{\rho}/(1-2K_{\rho})}. \quad (8)$$

Inserting this expression to eqs. (6) and (7), the excitation gap and the ground-state energy are given by $m = m(n) \propto V_{\perp}^{1/(2-4K_{\rho})}$ and $E = E(n) \propto V_{\perp}^{1/(1-2K_{\rho})}$. Thus, an *infinitesimal* value of interchain interaction transforms the system from TLL in the 1D case to the COI for *all* values of $0 < V \leq 2t$. T_{CO} can also be estimated in a similar way. The free energy up to n^2 is given by

$$f(n) = -\frac{\pi T^2}{6v} - \frac{b(K_{\rho})}{4\pi^2 v} \frac{z^2 V_{\perp}^2 n^2}{(2\pi T a/v)^{2-4K_{\rho}}} + \frac{z V_{\perp} n^2}{2a}, \quad (9)$$

where $b(K_{\rho}) \equiv c^2(K_{\rho}) \sin(2\pi K_{\rho}) B^2(K_{\rho}, 1 - 2K_{\rho})$ with $B(x, y) = \Gamma(x)\Gamma(y)/\Gamma(x+y)$. From the condition where the coefficient of n^2 in eq. (9) vanishes, T_{CO} is determined as

$$T_{\text{CO}} = \frac{v}{2\pi a} \left[\frac{z b(K_{\rho})}{2\pi^2 v} V_{\perp} a \right]^{1/(2-4K_{\rho})}. \quad (10)$$

At $V = 2t$, T_{CO} is proportional to V_{\perp} .^{16,17} In this case, the ratio of the gap at $T = 0$ and the transition temperature becomes $m/T_{\text{CO}}|_{V=2t} \approx 2.47$, which reproduces the result obtained in Ref. 23. The formulas of the amplitude n [eq. (8)] and the transition temperature T_{CO} [eq. (10)] are valid for $V \lesssim V_c^{1D}$, since these diverge when $V \rightarrow 0$ due to the unphysical ultraviolet divergence in eqs. (7) and (9). This problem could be resolved simply by introducing an ultraviolet cutoff.

Now we consider the case of finite U . An effective Hamiltonian for low-energy states ($k \approx \pm\pi/(4a)$) can be obtained by integrating out high-energy states.¹² The Hamiltonian is separated into the charge part describing the CO transition and the spin part. The latter is essentially the same as the Hamiltonian of 1D isotropic Heisenberg model and leads to gapless excitations both in the TLL and the COI. The density operator can be bosonized as

$$n_i = \frac{a}{\pi} \partial_x \theta_{\rho}(x) + \frac{2c_1}{\pi} \cos(2k_F x + \theta_{\rho}(x)) \sin \theta_{\sigma}(x) + c_2 \frac{(-1)^i}{\pi} \cos 2\theta_{\rho}(x), \quad (11)$$

where $\theta_{\rho}(x)$ and $\theta_{\sigma}(x)$ are the bosonic phase variables for the charge and spin degrees of freedom. c_1 and c_2 are nonuniversal numerical constants. In the absence of n , the charge part is written by the following phase Hamiltonian,

$$\mathcal{H}_{\rho} = \frac{v_{\rho}}{4\pi} \left[\frac{1}{K_{\rho 0}} (\partial_x \theta_{\rho})^2 + K_{\rho 0} (\partial_x \phi_{\rho})^2 \right] + \frac{g_{1/4}}{2(\pi a)^2} \cos 4\theta_{\rho}, \quad (12)$$

where $[\theta_{\rho}(x), -\partial_{x'} \phi_{\rho}(x')/(2\pi)] = i\delta(x - x')$. The charge velocity v_{ρ} , the parameter $K_{\rho 0}$, and the magnitude of the 1/4-filled Umklapp scattering $g_{1/4}$ are nonuniversal and its expressions obtained perturbatively are given in Ref. 12. In the presence of the interchain mean-field n , the additional Umklapp scattering appears, as in the last two terms in eq. (5),

$$\mathcal{H}' = \frac{zc_2}{\pi a} V_{\perp} n \cos 2\theta_{\rho} + \frac{z}{2a} V_{\perp} n^2. \quad (13)$$

By minimizing $\langle \mathcal{H}_{\rho} + \mathcal{H}' \rangle$ with respect to n , the amplitude n is determined as

$$n = -\frac{c_2}{\pi} \langle \cos 2\theta_{\rho} \rangle, \quad (14)$$

where $\langle \dots \rangle$ denotes the finite- T expectation value with respect to $\mathcal{H}_{\rho} + \mathcal{H}'$. We note that the parameter c_2 is proportional to the on-site interaction, $c_2 \approx U/(\sqrt{2}\pi t)$ within the

perturbative treatment. This implies that the CO fluctuation with 2-fold periodicity can emerge due to the correlation effect.

In order to estimate the second-order transition temperature, the lowest-order perturbation theory in n is sufficient. By expanding the r.h.s. of eq. (14) with respect to \mathcal{H}' , the equation to determine T_{CO} is obtained as¹⁷

$$1 = \frac{zc_2^2}{\pi^2 a} V_{\perp} \int dx \int_0^{1/T_{\text{CO}}} d\tau \times \langle T_{\tau} \cos 2\theta_{\rho}(x, \tau) \cos 2\theta_{\rho}(0, 0) \rangle_0 |_{T=T_{\text{CO}}}, \quad (15)$$

where $\langle \dots \rangle_0$ is the expectation value with respect to \mathcal{H}_{ρ} . In the following, we employ naive renormalization group (RG) arguments to estimate T_{CO} . By neglecting the anisotropy in space and time, the correlation function $\langle T_{\tau} \cos 2\theta_{\rho}(x, \tau) \cos 2\theta_{\rho}(0, 0) \rangle_0$ can be roughly calculated as

$$\langle T_{\tau} \cos 2\theta_{\rho}(x, \tau) \cos 2\theta_{\rho}(0, 0) \rangle_0 \approx \frac{1}{2} \exp \left[- \int_0^{\ln[\min(r_{\rho}, \xi_{\rho})/a]} dl (4K_{\rho}(l) + 2G_{1/4}(l)) \right], \quad (16)$$

where $r_{\rho} = \sqrt{x^2 + (v_{\rho}\tau)^2}$ and $\xi_{\rho} = v_{\rho}/(\pi T)$ is the characteristic thermal coherence length. The quantities $K_{\rho}(l)$ and $G_{1/4}(l)$ are the solution of the RG equations,

$$dK_{\rho}(l)/dl = -8G_{1/4}^2(l)K_{\rho}^2(l), \quad (17a)$$

$$dG_{1/4}(l)/dl = (2 - 8K_{\rho}(l))G_{1/4}(l), \quad (17b)$$

where the initial conditions are $K_{\rho}(0) = K_{\rho 0}$ and $G_{1/4}(0) = g_{1/4}/(2\pi v_{\rho})$. These RG equations have two kinds of fixed points for realistic parameters.¹² One is $(K_{\rho}(\infty), G_{1/4}(\infty)) = (0, -\infty)$ and the other is $(K_{\rho}(\infty), G_{1/4}(\infty)) = (K_{\rho}, 0)$. For $V_{\perp} = 0$ the former corresponds to the COI at $T = 0$, while the latter to the metallic TLL state. From eqs. (15) and (16), T_{CO} can be estimated by $F(bv_{\rho}/T_{\text{CO}}) = 1$ where

$$F(s) \equiv \frac{zc_2^2 a V_{\perp}}{\pi v_{\rho}} \int_0^{\ln s} dy \times \exp \left[- \int_0^y dl (4K_{\rho}(l) + 2G_{1/4}(l) - 2) \right], \quad (18)$$

and b is a positive numerical constant.

Concerning the realization of the COI at finite- T , we find three distinct regions on the (U, V) plane shown in Fig. 2. In region (iii) where the COI is already realized in the purely 1D case, infinitesimal V_{\perp} makes T_{CO} finite, as is naturally expected. In addition, there is a region (ii) with $1/2 \geq K_{\rho} \geq$

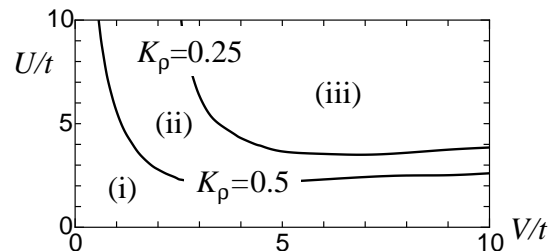


Fig. 2. Three distinct regions classified by appearance of the COI at the finite temperature.

Table I. The V_{\perp} dependence of the transition temperature T_{CO} in the respective regions, where A , B and C are positive constants. Corresponding K_{ρ} values are also listed. In the region (i), the transition happens only for $V_{\perp} > A(4K_{\rho} - 2)$.

	K_{ρ}	T_{CO}
(i)	$K_{\rho} \geq 1/2$	$\propto [1 - A(4K_{\rho} - 2)/V_{\perp}]^{1/(4K_{\rho}-2)}$
(ii)	$1/4 \leq K_{\rho} \leq 1/2$	$\propto [1 - A(4K_{\rho} - 2)/V_{\perp}]^{1/(4K_{\rho}-2)}$
(iii)	not defined	$= V_{\perp} [1 + B \ln(C/V_{\perp})]$

$1/4$ where *infinitesimal* V_{\perp} also gives rise to finite T_{CO} even in the metallic region for $V_{\perp} = 0$, since $F(\infty)$ diverges due to the divergence of CO fluctuation while $G_{1/4}(\infty) = 0$. This is consistent with the case of $U \rightarrow \infty$ discussed above, where $0 < V \leq 2t$ correspond to region (ii) in this limit, as mentioned. Moreover, even in the region (i) without divergence of CO fluctuation, *finite* amount of V_{\perp} makes the system COI. The obtained expressions for T_{CO} as a function of V_{\perp} is summarized in Table I. The critical value of V_{\perp} for appearance of the COI in the region (i) is proportional to $4K_{\rho} - 2$. We note that $T_{\text{CO}} \propto \exp(-A/V_{\perp})$ at $K_{\rho} = 1/2$, and $T_{\text{CO}} \propto V_{\perp}$ for $K_{\rho} = 1/4$, also consistent with the $U \rightarrow \infty$ case.

Now let us discuss the relevance of our results to the experiments. In $(\text{DI-DCNQI})_2\text{Ag}$, the non-metallic behavior above $T_{\text{CO}} \simeq 220 \text{ K}^3$ might be due to the $1/4$ -filled Umklapp scattering described in eq. (12). The peak at $T = T_{\text{CO}}$ in the derivative of the resistivity as a function of T in Ref. 3 seems to be triggered by the generation of additional Umklapp scattering $\cos 2\theta_{\rho}$ in eq. (13) due to the appearance of the CO. In $(\text{TMTTF})_2\text{X}$, the system is also insulating already above T_{CO} and a change in the slope of the resistivity curve is observed.²⁴ However, in this case the slight dimerization in the chain direction gives rise to another relevant $1/2$ -filled Umklapp scattering.¹² Whether or not the $1/4$ -filled Umklapp scattering is effective above T_{CO} is difficult to judge, but the change in the slope should be due to the additional Umklapp scattering by CO as discussed in this paper.

In these quasi 1D compounds, the interchain Coulomb interaction is in fact considered to be fairly large. For example in some members of $(\text{TMTTF})_2\text{X}$ it is even estimated to be comparable to the intrachain ones in quantum chemistry calculations.²⁵ This is noticeable since the interchain electron hopping is one order of magnitude smaller than that in the intrachain direction due to the anisotropic shape of the HOMO of the TMTTF molecule. However we should note also that the actual interchain networks are rather complicated. In the structure of $(\text{DCNQI})_2\text{X}$, it has a spiral symmetry when rounding each plaquette in the network. It is pointed out that this gives rise to frustration for the Wigner-crystal-type CO since the periodicity of the spiral does not fit the 2-fold period along the DCNQI chains.²⁶ In $(\text{TMTTF})_2\text{X}$, such frustration is more straightforwardly expected since the interchain coupling is zigzag-like therefore anti-phase and in-phase CO patterns would have close energy. These should reduce the effective interchain interaction to a smaller value at a first approximation.

In the present analysis, we considered the Wigner-crystal-type CO with a 2-fold periodicity only, namely, the “ $4k_{\text{F}}$ ” CDW state. In the small V region, however, the $2k_{\text{F}}$ CDW state having a 4-fold periodicity may also be stabilized, since its fluctuation develops as $\chi_c^{2k_{\text{F}}}(T) \propto T^{K_{\rho}-1}$. From the simple power counting, one finds that $2k_{\text{F}}$ fluctuation becomes

dominant if $K_{\rho} > 1/3$. Thus in the phase diagram where $1/3 < K_{\rho} < 1/2$, competition and/or coexistence of the CO and the $2k_{\text{F}}$ CDW may be possible. This problem needs further investigation.

In conclusion, we investigated the finite-temperature CO transition in the quasi-1D electron system at quarter filling coupled by the interchain Coulomb repulsion V_{\perp} , by using the interchain mean-field theory and the bosonization method. It was shown that the interchain interaction gives rise to finite transition temperature T_{CO} , with different behavior depending on the parameter in the 1D limit. When it is in the COI, V_{\perp} gives rise to finite temperature phase transition. In the TLL phase, in the parameter region where the charge-fluctuation develops, i.e., $1/2 > K_{\rho} \geq 1/4$, infinitesimally small V_{\perp} also produces the COI with finite T_{CO} , while for $K_{\rho} < 1/2$, at a finite critical value of V_{\perp} the system transform from the TLL to the COI.

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- 1) K. Hiraki and K. Kanoda: Phys. Rev. Lett. **80** (1998) 4737.
- 2) Y. Nogami *et al.*: J. Phys. IV France **9** (1999) Pr10-357.
- 3) T. Itou *et al.*: Phys. Rev. Lett. **93** (2004) 216408; Phys. Rev. B **72** (2005) 113109.
- 4) D.S. Chow *et al.*: Phys. Rev. Lett. **85** (2000) 1698; F. Zamborszky *et al.*: Phys. Rev. B **66** (2002) 081103; W. Yu *et al.*: *ibid.* **70** (2004) 121101.
- 5) P. Monceau, F. Ya. Nad and S. Brazovskii: Phys. Rev. Lett. **86** (2001) 4080.
- 6) T. Nakamura: J. Phys. Soc. Jpn. **72** (2003) 213.
- 7) For a review, H. Seo, C. Hotta, and H. Fukuyama: Chem. Rev. **104** (2004) 5005.
- 8) K. Sano and Y. Ōno: Phys. Rev. B **70** (2004) 155102; S. Ejima, F. Gebhard and S. Nishimoto: Europhys. Lett **70** (2005) 492 and references therein.
- 9) H. J. Schulz: in *Strongly Correlated Electronic Materials*, eds. K. S. Bedell, Z. Wang, D. E. Meltzer, A. V. Balatsky, and E. Abraham (Addison-Wesley Publish Company, 1994).
- 10) T. Giamarchi: Physica B **230-232** (1997) 975.
- 11) K. Yonemitsu: Phys. Rev. B **56** (1997) 7262.
- 12) H. Yoshioka, M. Tsuchiizu and Y. Suzumura: J. Phys. Soc. Jpn. **69** (2000) 651; **70** (2001) 762; M. Tsuchiizu, H. Yoshioka, and Y. Suzumura: *ibid.* **70** (2001) 1460.
- 13) C. Bourbonnais and L.G. Caron: Int. J. Mod. Phys. **5** (1991) 1033.
- 14) D. J. Scalapino, Y. Imry and P. Pincus: Phys. Rev. B **11** (1975) 2042.
- 15) A. A. Ovchinnikov: Sov. Phys. JETP **37** (1973) 176.
- 16) S. Inagaki and H. Fukuyama: J. Phys. Soc. Jpn. **57** (1988) 1435.
- 17) see Chap. 6.4.2 in T. Giamarchi: *Quantum Physics in One Dimension* (Oxford University Press, 2004).
- 18) H.J. Schulz: Phys. Rev. Lett. **77** (1996) 2790; Z. Wang: *ibid.* **78** (1997) 126; F.H.L. Essler, A.M. Tsvelik and G. Delfino: Phys. Rev. B **56** (1997) 11001; V.Y. Irkhin and A.A. Katanin: *ibid.* **61** (2000) 6757; M. Bocquet: *ibid.* **65** (2002) 184415.
- 19) M.J. Bhaseen and A.M. Tsvelik: cond-mat/0409602.
- 20) H.J. Schulz: Phys. Rev. Lett. **64** (1990) 2831.
- 21) S. Lukyanov: Phys. Rev. B **59** (1999) 11163.
- 22) A.I.B. Zamolodchikov: Int. J. Mod. Phys. A **10** (1995) 1125.
- 23) E. Orignac and R. Chitra: Phys. Rev. B **70** (2004) 214436.
- 24) C. Coulon *et al.*: J. Physique **43** (1982) 1059.
- 25) A. Fritsch and L. Ducasse: J. Phys. I France **1** (1991) 855; F. Castet, A. Fritsch, and L. Ducasse: *ibid.* **6** (1996) 583.
- 26) K. Kanoda *et al.*: J. Phys. IV France **131** (2005) 21.